Supplementary Information

Ferroelectric Rashba semiconductors $AgBiP_2X_6$ (X = S, Se and Te) with valley polarization: An avenue towards electric and nonvolatile control of spintronic devices

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Table S1. The calculated lattice constant *a*, thickness *d*, bond length, and displacement of Ag^{+} and Bi^{3+} ions from the centrosymmetric positions.

		$AgBiP_2S_6$	$AgBiP_2Se_6$	$AgBiP_2Te_6$
a (Å)		6.446	6.745	7.189
d (Å)		3.441	3.586	3.724
P-P bonds		2.263	2.285	2.302
Ag-X bonds	long	3.061	3.071	3.363
	short	2.650	2.792	2.866
Bi-X bonds	long	2.878	2.991	3.201
	short	2.853	2.975	3.128
Ag^{+} displacement		0.487	0.428	0.833
Bi^{3+} displacement		0.168	0.194	0.364

Table S2. The lattice vectors and fractional coordinates of each atom in $AgBiP_2S_6$ monolayer.

a [Å]	6.4460170697170192	0.0000000000000000	0.0000000000000000
b [Å]	-3.2230085334649878	5.5824145364076143	0.0000000000000000
c [Å]	0.0000000000000000	0.0000000000000000	23.5874996185000008
S	0.9779085968483583	0.6256006066924290	0.4951239831492113
S	0.3743993933075708	0.3523079901559291	0.4951239831492113
S	0.6476920098440710	0.0220914031516418	0.4951239831492113
S	0.3491938134844143	0.0456602013067838	0.3492060319203195
S	0.9543397986932159	0.3035336121776305	0.3492060319203195
S	0.6964663878223694	0.6508061865155855	0.3492060319203195
P	0.6666666666666643	0.3333333333333357	0.3727974718048547
P	0.6666666666666643	0.3333333333333357	0.4687315826592956
Ag	0.0000000000000000	0.0000000000000000	0.4428369518611184
Bi	0.3333333333333357	0.6666666666666643	0.4150239814661507

Table S3. The lattice vectors and fractional coordinates of each atom in $AgBiP_2Se_6$ monolayer.

a [Å]	6.7451425251032466	0.0000000000000000	0.0000000000000000
b [Å]	-3.3725712622740640	5.8414647790463770	0.0000000000000000
c [Å]	0.0000000000000000	0.0000000000000000	23.5874996185000008
Se	0.9899088918095423	0.6339494198664021	0.4986792991742209
Se	0.3660505801335979	0.3559594719431403	0.4986792991742209
Se	0.6440405280568597	0.0100911081904575	0.4986792991742209
Se	0.3329223877526660	0.0411902968239272	0.3466420743351128
Se	0.9588097031760727	0.2917320909287389	0.3466420743351128
Se	0.7082679090712611	0.6670776122473341	0.3466420743351128
P	0.6666666666666643	0.3333333333333357	0.3721448632042433
P	0.6666666666666643	0.3333333333333357	0.4690332653703307
Ag	0.0000000000000000	0.0000000000000000	0.4408164250831848
Bi	0.3333333333333357	0.6666666666666643	0.4144213588142523

Table S4. The lattice vectors and fractional coordinates of each atom in $AgBiP_2Te_6$ monolayer.

a [Å]	7.1894800363574678	0.0000000000000000	0.0000000000000000
b [Å]	-3.5947399253376240	6.2262724050884772	0.0000000000000000
c [Å]	0.0000000000000000	0.0000000000000000	23.5874996185000008
Te	0.0130186388505032	0.6345696391814084	0.5014714336207148
Te	0.3654303608185918	0.3784489996690950	0.5014714336207148
Te	0.6215510003309047	-0.0130186388505031	0.5014714336207148
Te	0.2981748686531853	0.0414301448793267	0.3435898436919037
Te	0.9585698551206728	0.2567447237738584	0.3435898436919037
Te	0.7432552762261413	0.7018251313468150	0.3435898436919037
P	0.6666666666666643	0.3333333333333357	0.3673332899681562
P	0.6666666666666643	0.3333333333333357	0.4649091420753992
Ag	0.0000000000000000	0.0000000000000000	0.4578673389448687
Bi	0.3333333333333357	0.6666666666666643	0.4070864300737347

Table S5. The calculated Rashba energy (E_R), Rashba parameter (α_R), and band-gap (E_g) under different biaxial strain.

	E_R (meV)	α_R (eVÅ)	E_g (eV)
ε = +8%	1.3	0.9	0.82
ε = +6%	1.8	1.7	0.69
ε = +4%	2.5	3.1	0.52
ε = +2%	4.9	6.1	0.50
ε = 0%	6.5	6.5	0.49
ε = –2%	7.7	6.8	0.48
ε = –4%	8.9	7.2	0.38
ε = –6%	5.9	4.7	0.24
ε = –8%	—	—	0.02

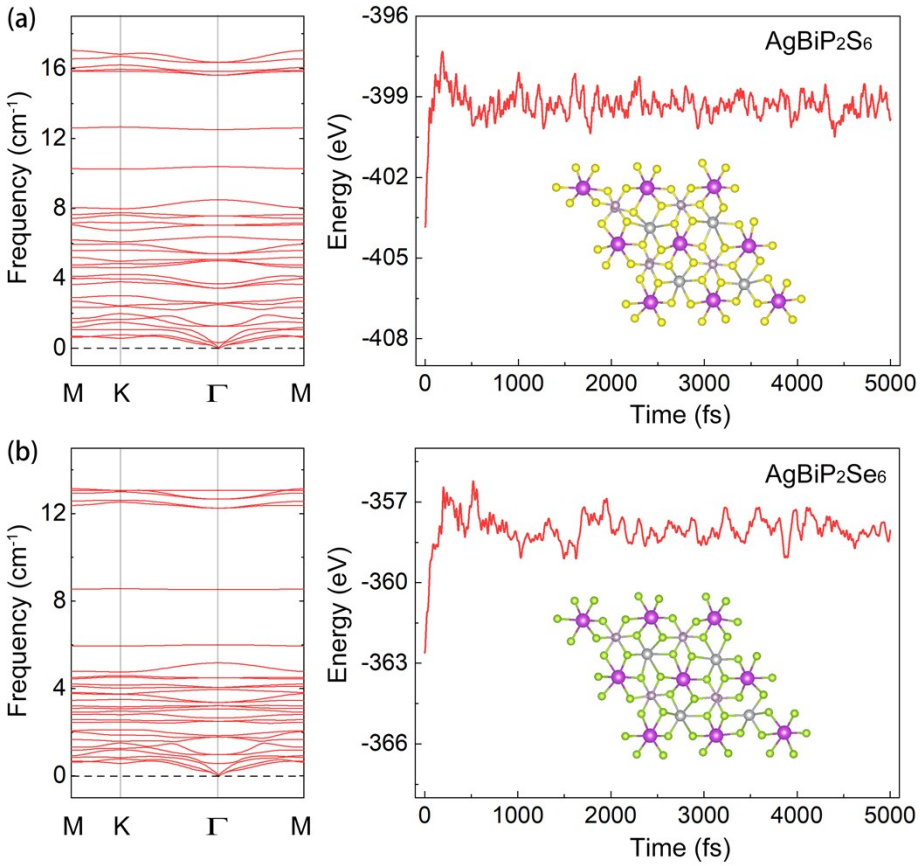

Fig. S1. The phonon band dispersions and potential energy fluctuations of (a) $AgBiP_2S_6$ and (b) $AgBiP_2Se_6$ monolayers. The inset shows the corresponding structure at 300 K after the simulation for 5 ps.

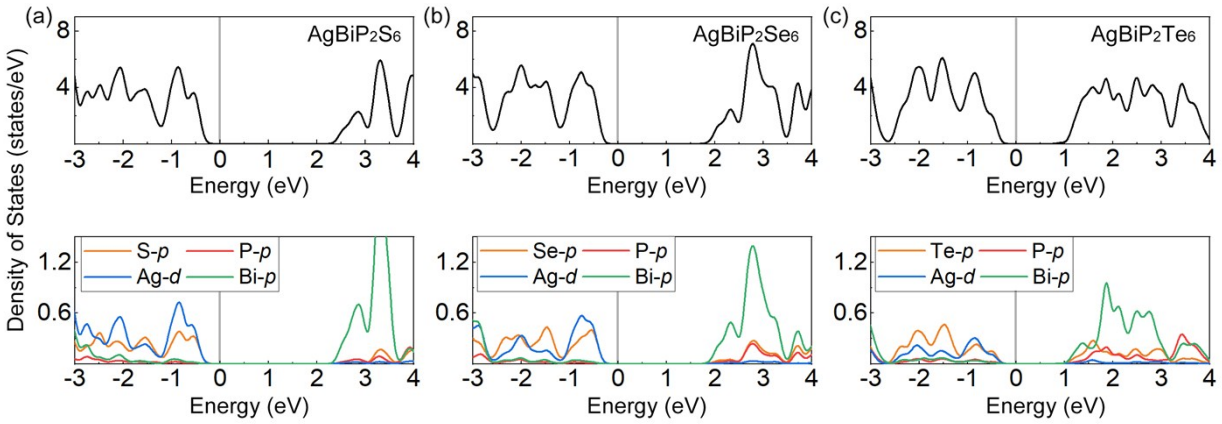

Fig. S2. The total DOS and orbital-resolved PDOS for each atom of $AgBiP_2X_6$ monolayers calculated by HSE06 method.

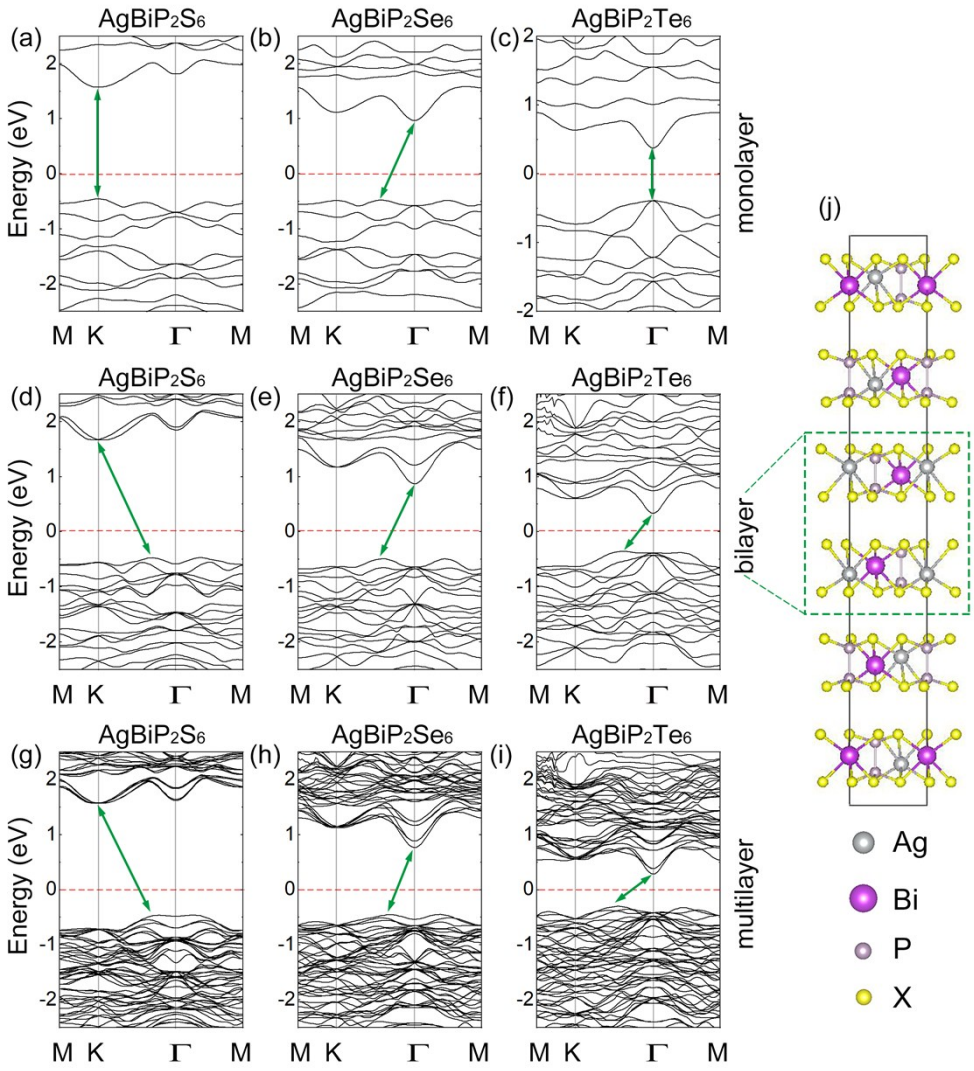

Fig. S3. Calculated band structures of (a-c) $AgBiP_2X_6$ monolayers, (d-f) $AgBiP_2X_6$ bilayers, and (g-i) $AgBiP_2X_6$ multilayers by PBE method. The Fermi level is set to zero. (j) The structure of $AgBiP_2X_6$ multilayer with interlayer anti-ferroelectric.

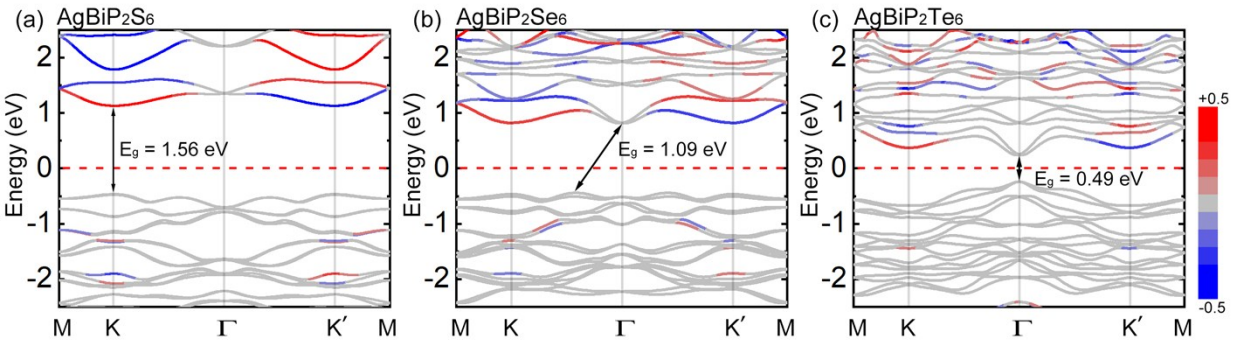

Fig. S4. Calculated band structures of $AgBiP_2X_6$ monolayers with SOC. The spin projections of Bi-*p* orbitals along *z* direction are represented by red and blue lines, which represent the spin-up and spin-down states, respectively. The Fermi level is set to zero.

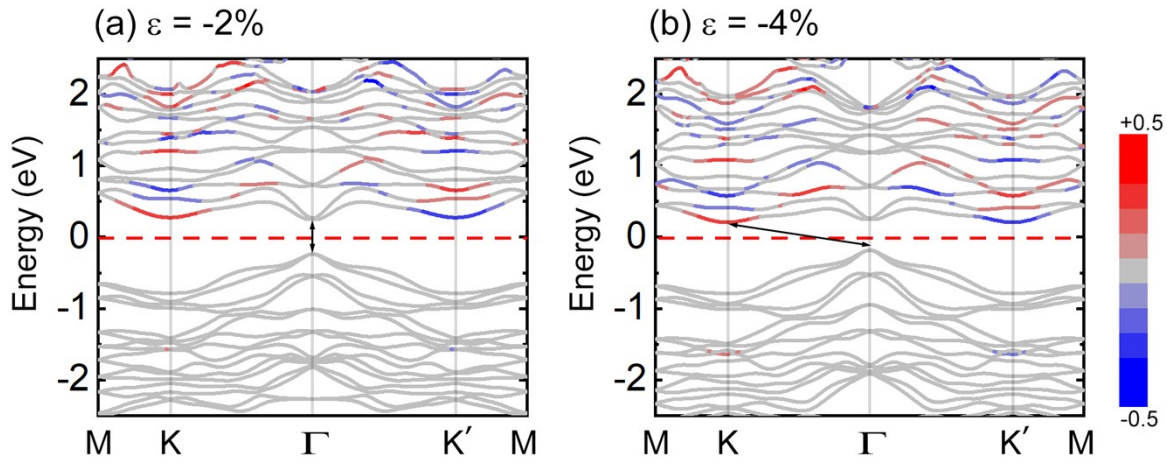

Fig. S5. Calculated band structures of compressed $AgBiP_2Te_6$ monolayers with SOC. (a) ε = -2%. (b) ε = -4%. The spin projections of Bi-*p* orbitals along *z* direction are represented by red and blue lines, which represent the spin-up and spin-down states, respectively. The Fermi level is set to zero.